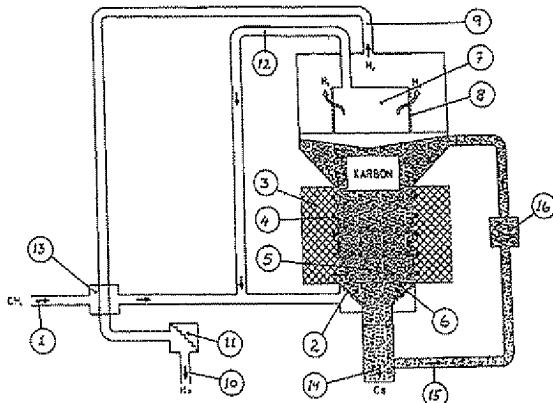


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(54) PRODUCTION D'HYDROGÈNE ET DE CARBONE A L'AIDE D'UN CATALYSEUR AU NOIR DE CARBONE  
 (54) PRODUCTION OF HYDROGEN AND CARBON WITH A CARBON BLACK CATALYST

(57) The invention covers method, device and application of production of hydrogen and carbon by pyrolysis based on natural gas, methane or other organic gases as raw material. The method for precipitation of solid carbon is characterized by the use of finely distributed carbon dust as catalyst for the precipitation process. The device is designed as a reaction chamber that contains the catalyst. The temperature in the chamber is controlled by supply of electrical power or other energy. In addition the invention covers the application of compact pyrolysis systems in vehicles, for pre-processing of gases containing hydrocarbons and for fuel production for polymer fuel cells that generate electrical power for propulsion of the vehicle.



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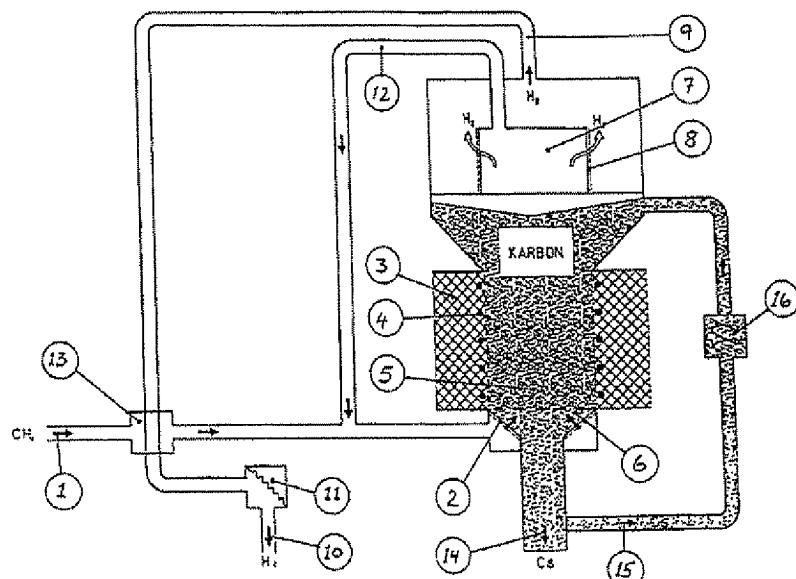
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<p>(21) International Application Number: <b>PCT/NO99/00299</b></p> <p>(22) International Filing Date: <b>30 September 1999 (30.09.99)</b></p> <p>(30) Priority Data: 19984560                    30 September 1998 (30.09.98) NO</p> <p>(71) Applicant (<i>for all designated States except US</i>): PROTOTECH AS [NO/NO]; Fantoftvegen 38, P.O. Box 6034, Postterminalen, N-5892 Bergen (NO).</p> <p>(72) Inventor; and</p> <p>(75) Inventor/Applicant (<i>for US only</i>): ARILD, Vik [NO/NO]; Slottenvien 76, N-5258 Blomsterdalen (NO).</p> <p>(74) Agent: CHRISTIAN MICHELSEN RESEARCH AS; Fantoftvegen 38, P.O. Box 6031, Postterminalen, N-5892 Bergen (NO).</p>		<p>(81) Designated States: AL, AM, AU, AZ, BA, BG, BR, BY, CA, CN, CU, CZ, DE, EE, GE, HR, HU, ID, IL, IN, IS, KG, KP, KR, KZ, LT, LV, MK, MX, NZ, PL, RO, RU, SI, SK, TJ, TM, UA, US, UZ, YU, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).</p> <p><b>Published</b>  <i>With international search report.</i>  <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>  <i>In English translation (filed in Norwegian).</i> </p>	
<p>(54) Title: PRODUCTION OF HYDROGEN AND CARBON WITH A CARBON BLACK CATALYST</p>			
<p>(57) Abstract</p> <p>The invention covers method, device and application of production of hydrogen and carbon by pyrolysis based on natural gas, methane or other organic gases as raw material. The method for precipitation of solid carbon is characterized by the use of finely distributed carbon dust as catalyst for the precipitation process. The device is designed as a reaction chamber that contains the catalyst. The temperature in the chamber is controlled by supply of electrical power or other energy. In addition the invention covers the application of compact pyrolysis systems in vehicles, for pre-processing of gases containing hydrocarbons and for fuel production for polymer fuel cells that generate electrical power for propulsion of the vehicle.</p>			

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## 1

## PRODUCTION OF HYDROGEN AND CARBON WITH A CARBON BLACK CATALYST.

The invention includes method, device and application of energy efficient production of hydrogen and carbon by pyrolysis based on natural gas, methane or other organic gases as raw material. The method for precipitation of solid carbon is characterised by the use of finely distributed carbon dust as catalyst for the precipitation process. Carbon molecules from the gas attach to the catalytic particles causing growth of these to a trappable size. The catalytic material is regenerated by continuous supply of finely crushed carbon from the process.

The device is designed as a heat insulated reaction chamber with room for the catalytic material. The temperature in the reaction zone is controlled by means of supplied energy. Heating can also take place using alternative heat sources, and the system may therefore use excessive heat from high temperature processes as energy source for the complete- or parts of the process. The device has shown good efficiency in a temperature range from 400°C to 2000°C. The reaction rate and the purity of the final products can be controlled by optimisation of pressure and temperature.

In addition the invention covers the application of compact pyrolysis systems for use in vehicles, for pre-processing of gasses containing hydrocarbons and for fuel production for polymer fuel cells. The fuel cells utilise hydrogen as fuel and generate electrical power for propulsion of the vehicle. Both the pyrolysis system and the fuel cells can be designed compactly to fit ordinary vehicles.

The device and method is particularly well suited in environments with limited supply of hydrogen and oxygen, but with good energy supplies. An example of such environments is the vehicles and units that operate outside the earth's atmosphere.

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Chemically clean carbon (carbon black) has been an important industrial product for many years. Large quantities are used in the production of car tires. The material is also used in paint products, in lubricants and in medical products. A number of methods for production of carbon from hydrocarbon gases have been developed during a period of years. Splitting of carbon and hydrogen from such gases is currently in focus from environmental reasons in connection with natural gas based production of electrical power. Also the space industry has interest in the hydrogen production as part of the water production in manned space journeys/stations.

A known method for splitting of hydrocarbons is the use of plasma arc. This method is described in US.Pat.no. 5,527,518. Another method is described in US.Pat.no. 4,631,180. Both methods involve combustion and use oxygen in the production.

A method for splitting of hydrocarbons is described in US.Pat.no. 5,198,084. This method is used for gasification of carbon containing material, and the gas is heated by means of microwave technology in a so-called plasma reactor.

The referred methods for splitting of hydrogen and carbon from hydrocarbons utilise different heating and combustion processes in atmospheres with insufficient oxygen supply. The method according to the invention significantly differs from these techniques by utilising carbon dust as catalyst for splitting of hydrocarbons in an oxygen free environment.

A patent DD 118263 describes a method for pyrolysis where the carbon particles are used as catalyst. The particles are sent through a gas containing hydrogen which is heated to a temperature of 1000°C - 1800°C. The invention differs significantly from this by the fact that device and method is based on

stationary carbon particles contained in a compact reaction chamber. This make it possible to produce a much more compact system compared to systems with moving particles or carbon deposition on surfaces. In addition, the new method is significantly more energy effective because the pyrolysis process operates at temperatures down to 400°C.

The method and device, according to the invention, are to be used in a process system for production of hydrogen and carbon based on natural gas, methane or other organic gases as raw material. The system is shown in principle-sketch fig.1. Gas (1) containing hydro carbons is guided through a filter (2), into a heat insulated reaction chamber (3) and heated by means of electrical heating coils or excessive heat from other high temperature processes. The temperature in the reaction chamber (3) is given an increasing gradient in the direction of flow (from bottom to top) from 300 to maximum 2000°C. The reaction chamber (3) contains finely distributed carbon dust (5) that acts as catalyst for the collection of solid carbon from the gas. The carbon molecules in the heated gas attach to the carbon dust (5) in a way that causes the catalytic particles to grow. The growing carbon particles are trapped by means of a mechanical system (for example a centrifuge) in the lower parts of the reaction chamber (6), when the grain size reaches a certain level. The carbon content in the gas gets a decreasing gradient upwards in the reaction chamber (3), and the gas contains mainly hydrogen at the top (12). The hydrogen-enriched gas is guided to a separation chamber (7), where parts of the gas are separated through a membrane filter (8). The permeate fraction of the gas (9) can be optimised with regard to the purity of the hydrogen. Before storage (10) the gas is guided through a filter (11) for removal of trace constituents. The retentate fraction of the gas (12) from the separation chamber (7) is returned to the inlet side of the reaction chamber.

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On its way to the trace constituents filter (11) the processed gas (9) passes through a heat exchanger (13) for pre-heating of the feed gas (1). The exchange of heat between processed and feed gas induces a reduction in the need for energy supply to the system.

Trapping of granulated carbon takes place continuously in the lower parts of the reaction chamber (6). As the catalytic particles grow and get trapped the system needs supply of new catalytic material. According to the invention, catalytic material is continuously produced by recycling, crushing (16) and injection in the upper part of the reception chamber of a controlled fraction (15) of the separated carbon (14). This recycling process maintains an optimum balance with regard to the amount and size distribution of carbon particles.

## Patent claims

1. Method for production of hydrogen and carbon by pyrolysis of methane and other organic gases utilising carbon dust as catalyst for precipitation of carbon in a closed process *characterised by* the stimulation of carbon precipitation by guiding the gas through a heated reaction chamber where the carbon molecules from the gas can attach to the catalytic particles causing growth of these to a pre-set size that can be mechanically trapped.
2. Method for production of hydrogen and carbon by pyrolysis of methane and other organic gases according to claim 1 and 2 *characterised by* the crushing of a controllable amount of precipitated carbon and the return of this to the reaction chamber in a continuous process for maintenance of an optimum balance with regard to the amount and size distribution of carbon particles.
3. Device for production of hydrogen and carbon by pyrolysis of methane and other organic gases in a closed system with a heat insulated reaction chamber *characterised by* the filling of the chamber with porous carbon dust with catalytic character and the temperature control by supply of electric power or excessive heat from high temperature processes.
4. The application of compact pyrolysis systems in vehicles for pre-processing of natural gas, methane and other organic gases with the aim of producing hydrogen fuel for the polymer fuel cells that generates electrical power for propulsion of the vehicle.

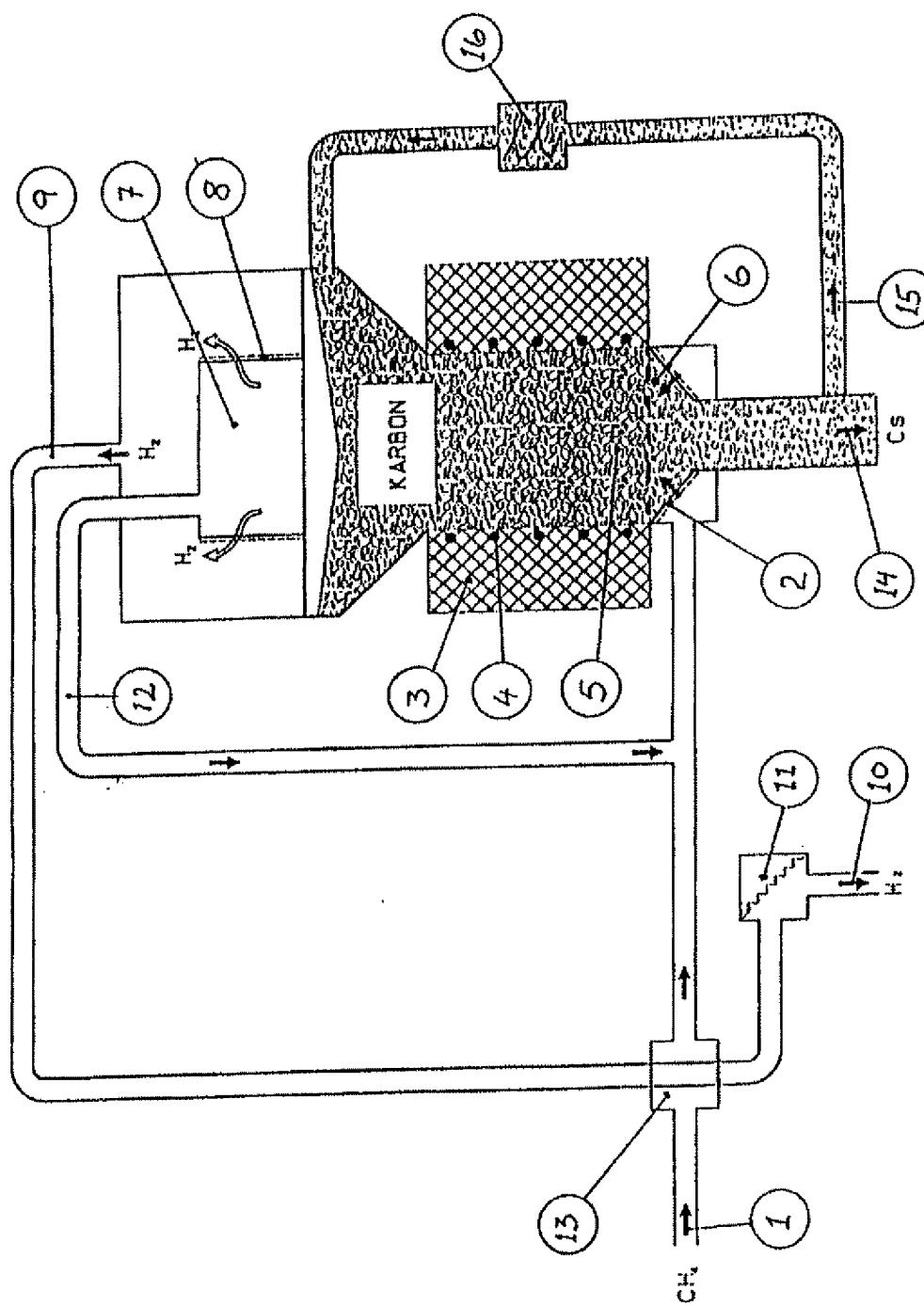


Fig. 1